

Disorder effects in diluted ferromagnetic semiconductors.

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Carrier induced ferromagnetism in diluted III-V semi-conductor is analyzed within a two step approach. First, within a single site CPA formalism, we calculate the element resolved averaged Green's function of the itinerant carrier. Then using a generalized RKKY formula we evaluate the Mn-Mn long-range exchange integrals and the Curie temperature as a function of the exchange parameter, magnetic impurity concentration and carrier density. The effect of the disorder (impurity scattering) appears to play a crucial role. The standard RKKY calculation (no scattering processes), strongly underestimate the Curie temperature and is inappropriate to describe magnetism in diluted magnetic semi-conductors. It is also shown that an antiferromagnetic exchange favors higher Curie temperature.

PACS numbers: 75.10.-b, 71.10.-w, 75.50.Dd

After the recent discovery by Ohno et al. that by doping GaAs [1] with only 5% of magnetic impurities Mn^{2+} , the Curie temperature T_C could already exceed 100 K and because of all the possible technological applications, the interest for the III-V diluted magnetic semiconductors (DMS) has increased considerably. In spite of the apparent success of different methods (mean field, first-principle, random-phase approximation (RPA)) where disorder is either neglected or treated at the lowest order to reproduce the Curie temperature [2–8], there is still some shadow region concerning the effect of disorder on magnetism. Indeed only few works, mainly based on numerical simulations, are including the effect of positional disorder [9,10] within a mean field treatment (multi-scattering effects are not included). Recently, in order to provide a simultaneous and self-consistent treatment of the itinerant carrier and magnetic impurity an approach based on the equation of motion method was proposed [8]. However, as a consequence of the RPA decoupling the self-energy of the itinerant carriers Green's function (GF) is reduced to the lowest order term $\Sigma_\sigma = \frac{z_\sigma}{2} J_{pd} c \langle \hat{S}^z \rangle$ (where $z_\sigma = \pm 1$ and c is Mn^{2+} concentration). Because of the difficulty to include within this formalism, higher order scattering terms, we follow in this paper a slightly different approach and focus first on the effect of the disorder on the itinerant carriers. First, we calculate the itinerant carrier GF by treating the effect of disorder in the full coherent-potential approximation (CPA) approximation, which means that all single site multi-scattering processes are properly included. In the second step we calculate the exchange integrals between spin impurities using the projected GF on the Mn sites. The difficulty is to perform properly the averaged T-matrix calculation since the holes/electrons scattering depends on the impurity spin operator. For that purpose we follow the procedure described in [11,12]. It should be added that in this work, spin impurities are treated fully quantum mechanically.

We consider the following minimal Hamiltonian which

is the good starting point to study DMS,

$$H = \sum_{ij,\sigma} t_{ij} c_{i\sigma}^\dagger c_{j\sigma} + \sum_i J_i \mathbf{S}_i \cdot \mathbf{s}_i \quad (1)$$

$t_{ij} = t$ for i and j nearest neighbors and zero otherwise. In the exchange between localized impurities spin and itinerant electron gas J_i is a random variable: $J_i = J$ ($J \geq 0$ means antiferromagnetic coupling) if the site i is occupied by a magnetic impurity, and zero otherwise. \mathbf{S}_i is the magnetic impurity spin operator at site i and $\mathbf{s}_i = c_{i\alpha}^\dagger (1/2 \boldsymbol{\sigma}_{\alpha\beta}) c_{i\beta}$ is the spin operator at site i of the itinerant electron gas.

The T-matrix associated to the multi-scattering of a single magnetic impurity (at site m) embedded in the effective medium is,

$$\hat{\mathbf{t}}_m = \hat{\mathbf{V}}_m (\mathbf{1} - \hat{\mathbf{G}} \hat{\mathbf{V}}_m)^{-1} \quad (2)$$

where,

$$\hat{\mathbf{V}}_m^{\text{Mn}} = \begin{pmatrix} \frac{1}{2} J S_m^z - \Sigma_\uparrow & \frac{1}{2} J S_m^- \\ \frac{1}{2} J S_m^+ & -\frac{1}{2} J S_m^z - \Sigma_\downarrow \end{pmatrix} \quad (3)$$

Respectively, for Ga at site m , $\hat{\mathbf{V}}_m^{\text{Ga}}$ is obtained by taking $J = 0$ in the previous equation.

The 2×2 averaged Green's function matrix $\hat{\mathbf{G}}$ is,

$$\hat{\mathbf{G}} = \begin{pmatrix} \hat{\mathbf{G}}_\uparrow & 0 \\ 0 & \hat{\mathbf{G}}_\downarrow \end{pmatrix} \quad (4)$$

with $\hat{\mathbf{G}}_\sigma = (\omega \mathbf{I} - \hat{\mathbf{K}}_\sigma)^{-1}$ where, $\hat{\mathbf{K}}_\sigma = \sum_{\mathbf{k}} (\epsilon_{\mathbf{k}} - \Sigma_\sigma) c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma}$.

In the following, we omit the site index m . The self-energy Σ_σ is obtained by solving the coupled self-consistent equations.

$$\langle t_{\sigma\sigma} \rangle_{\text{dis},T} = (1 - c) \langle t_{\sigma\sigma}^{\text{Ga}} \rangle_T + c \langle t_{\sigma\sigma}^{\text{Mn}} \rangle_T = 0 \quad (5)$$

$\langle \dots \rangle_{\text{dis}, T}$ denotes configuration average and thermal average at temperature T for the spin operator, c is the concentration of Mn impurities.

After lengthy calculations we get,

$$\Sigma_\sigma = \frac{\langle \sum_\lambda x_\lambda \hat{\mathbf{c}}_\sigma^\lambda (\hat{\mathbf{d}}_\sigma^\lambda)^{-1} \rangle_T}{\langle \sum_\lambda x_\lambda (\hat{\mathbf{d}}_\sigma^\lambda)^{-1} \rangle_T} \quad (6)$$

The sum runs over all constituents, in our binary system $x_{\text{Mn}} = c$ (resp. $x_{\text{Ga}} = 1 - c$).

$$\begin{aligned} \hat{\mathbf{c}}_\sigma^{\text{Mn}} = & z_\sigma \frac{J}{2} \hat{\mathbf{S}}^z + \left(\frac{J^2}{4} [S(S+1) - (\hat{\mathbf{S}}^z)^2 - z_\sigma \hat{\mathbf{S}}^z] \right. \\ & \left. + [z_\sigma \frac{J}{2} \hat{\mathbf{S}}^z + \Sigma_\sigma] \left[\frac{J}{2} (z_{-\sigma} \hat{\mathbf{S}}^z - 1) - \Sigma_{-\sigma} \right] G_{-\sigma} \right) \end{aligned} \quad (7)$$

and,

$$\begin{aligned} \hat{\mathbf{d}}_\sigma^{\text{Mn}} = & 1 - G_{-\sigma} \left[\frac{J}{2} (z_{-\sigma} \hat{\mathbf{S}}^z - 1) - \Sigma_{-\sigma} \right] \\ & - G_\sigma [\hat{\mathbf{c}}_\sigma^{\text{Mn}} - z_\sigma \frac{J}{2} \hat{\mathbf{S}}^z] \end{aligned} \quad (8)$$

with $z_\sigma = 1$ (resp. -1) for spin \uparrow (resp. \downarrow). $\bar{G}_\sigma = \frac{1}{N} \sum_{\mathbf{q}} \bar{G}_\sigma(\mathbf{q}, \omega)$ where $\bar{G}_\sigma(\mathbf{q}, \omega) = (\omega - \epsilon_{\mathbf{q}} - \Sigma_\sigma(\omega))^{-1}$ denotes the averaged GF. Similarly, one gets $\hat{\mathbf{c}}_\sigma^{\text{Ga}}$ and $\hat{\mathbf{d}}_\sigma^{\text{Ga}}$ after setting $J = 0$ in the previous equations.

Note that since impurity spins are treated *quantum mechanically*, the thermal averaged quantities are evaluated using the following decomposition, $\langle \hat{\mathbf{O}}(\hat{\mathbf{S}}^z) \rangle_T = \sum_{i=0}^{2S} a_i \langle (\hat{\mathbf{S}}^z)^i \rangle_T$ where $\hat{\mathbf{O}}$ denotes a general operator which depends in a non trivial manner on $(\hat{\mathbf{S}}^z)^i$. Additionally, as it was shown by Callen-Strikman, $\langle (\hat{\mathbf{S}}^z)^i \rangle_T$, and hence $\langle \hat{\mathbf{O}}(\hat{\mathbf{S}}^z) \rangle_T$, are universal functions of $\langle \hat{\mathbf{S}}^z \rangle_T$ only [13]. After solving the coupled set of equations (Eq. (6) with $\sigma = \pm 1$) one gets the total averaged GF of the itinerant carriers.

The next step consists in calculating the long-range exchange integrals J_{ij}^{eff} between magnetic impurities for the effective Heisenberg Hamiltonian,

$$H^{\text{Heis}} = \frac{1}{2} \sum_{i \neq j} J_{ij}^{\text{eff}} \mathbf{S}_i \cdot \mathbf{S}_j \quad (9)$$

The exchange integrals between two impurities separated by a distance \mathbf{R} is given by the generalized RKKY formula,

$$J^{\text{eff}}(\mathbf{R}) = -\frac{1}{2} J^2 \left[-\frac{1}{\pi} \text{Im} \chi(\mathbf{R}) \right] \quad (10)$$

where the susceptibility is,

$$\chi(\mathbf{R}) = \sum_{\mathbf{k}, \mathbf{q}} \int d\omega f(\omega) \bar{G}_\uparrow^{\text{Mn}}(\mathbf{k}, \omega) \bar{G}_\downarrow^{\text{Mn}}(\mathbf{k} + \mathbf{q}, \omega) e^{i\mathbf{q} \cdot \mathbf{R}} \quad (11)$$

The chemical potential μ entering the Fermi-Dirac function $f(\omega)$ is determined at each temperature by fixing

the itinerant carrier density. Note that the exchange integrals are T -dependent through the averaged GF. When replacing \bar{G}_σ by the free particle GF the exchange integrals reduce to the standard RKKY. Additionally, it is important to stress that to calculate J_{ij}^{eff} , one has to take into account that both site i and j should be occupied by Mn atom. Thus the non local GF which enters Eq. 11 should be the Mn-resolved GF but not the full averaged one.

To derive the projected GF on Mn sites, we essentially follow the procedure described in Ref. [14], which gives,

$$\begin{aligned} \bar{G}_\sigma^{\text{Mn}}(\mathbf{k}, \omega) = & F_\sigma(\omega) (1 - F_\sigma(\omega)) \bar{G}_\sigma(\omega) \\ & + F_\sigma^2(\omega) \bar{G}_\sigma(\mathbf{k}, \omega) \end{aligned} \quad (12)$$

where $\bar{G}_\sigma(\omega) = \frac{1}{N} \sum_{\mathbf{k}} \bar{G}_\sigma(\mathbf{k}, \omega)$ and $F_\sigma(\omega) = \left(1 - \bar{G}_\sigma(\omega) (V_{\text{eff}, \sigma}^{\text{Mn}} - \Sigma_\sigma) \right)^{-1}$. The determination of the effective potential $V_{\text{eff}, \sigma}^{\text{Mn}}$ leads to,

$$V_{\text{eff}, \sigma}^{\text{Mn}} = \Sigma_\sigma \frac{1 + \Sigma_\sigma \bar{G}_\sigma}{c + \Sigma_\sigma \bar{G}_\sigma} \quad (13)$$

As a final step we can evaluate the Curie temperature, by using mean-field theory for the effective Heisenberg model:

$$k_B T_C = \frac{2}{3} S(S+1) c \frac{1}{N} \sum_{\mathbf{q}} E(\mathbf{q}) \quad (14)$$

$E(\mathbf{q})$ is the T -dependent magnon spectrum: $E(\mathbf{q}) = \tilde{J}^{\text{eff}}(\mathbf{0}) - \tilde{J}^{\text{eff}}(\mathbf{q})$ where $\tilde{J}^{\text{eff}}(\mathbf{q})$ denotes the Fourier transform of the exchange integrals.

In the following we discuss the numerical results. In Fig. 1 we have plotted the total density of states (DOS) and the projected one on Mn site as a function of energy, for different values of the parameter J/t . We observe that in the weak coupling regime ($J/t = 0.86$) the total spin-resolved DOS is almost identical to the unperturbed one although the Mn-projected DOS is already strongly affected by disorder. By increasing further J/t , we observe at low energy the impurity band formation. Note that the impurity band splits first at $E \leq 0$, and the position of the peaks in the Mn-DOS are not symmetric with respect to 0. This can be easily understood by analyzing the atomic limit ($J/t \rightarrow \infty$) which is properly described. In the paramagnetic phase, we get a peak at $E_{\text{high}} = +\frac{1}{2}JS$ and another at $E_{\text{low}} = -\frac{1}{2}J(S+1)$ with respective weights $\frac{S+1}{2S+1}c$ and $\frac{S}{2S+1}c$ [16].

In Fig. 2, the dependence of T_C on $\gamma = n_h/c$ (n_h is the hole concentration) is discussed. At fixed γ , the Curie temperature increases significantly with J/t and large value are reached when approaching the split-band regime. In the intermediate regime ($J/t \geq 2$), T_c appears to be *very sensitive* to J/t , a maximum at $\gamma \approx 0.10$ is observed before T_C decrease and eventually vanishes at γ_c which is J/t dependent. These results are qualitatively comparable to those of ref. [15], although we obtain Curie

temperature significantly larger. Additionally, in comparable regime the maximum of T_C in ref. [15] corresponds to half-filled impurity band ($\gamma \approx 0.50$) and T_C is symmetric with respect to this point (it vanishes at $\gamma = 1$). Later it will be shown that the sign of J which is irrelevant in most of the model calculations, plays in fact an important role. As it will be discussed in the next section, the value of γ_c for which T_C vanishes depends on both the sign and amplitude of J/t .

In Fig. 3, T_C as a function of J/t is shown for both antiferromagnetic and ferromagnetic coupling for different carrier density. First, the sign of J/t appears to be relevant. Indeed, T_C is strongly asymmetric with respect to $J/t = 0$. In the case of ferromagnetic coupling the maximum of T_C is *much smaller* than for antiferromagnetic coupling. However, as expected, for $|J|/t \leq 1$ they are comparable and reduce to the standard RKKY calculations ($T_C \propto J^2$). For both, ferromagnetic and antiferromagnetic coupling, the position of the maximum depends on the hole concentration. However, the maximum occurs earlier in the ferromagnetic case. Note that in the *intermediate regime* $1 \leq J/t \leq 3$, T_C^{RKKY} is *much smaller* than T_C^{CPA} : for $J/t = 2$, $T_C^{\text{CPA}} \approx 3 T_C^{\text{RKKY}}$ for $n_h = 0.015$. Additionally, after the maximum is reached, T_C drops rapidly, and vanishes at a n_h -dependent value of J/t . We observe that in the split-band regime $J/t \geq 3.5$ *no ferromagnetic ordering* is possible. In contrast to other approaches, our theory appears to be more suitable to describe the large coupling regime.

Let us discuss briefly, the relevance of our results with respect to experimental data of $\text{Ga}_c\text{Mn}_{1-c}\text{As}$. Our model is based on a one-band model, as done in Ref. [8], we fix t by assuming a hole effective mass $m^* = 0.5 m_e$. This leads to a value $t = 0.58$ eV [17]. We assume that the 5.3%-doped sample (highest $T_C = 110$ K) contains $n_h \approx 0.2 c$. Using Fig.4, we obtain $J = -1.12$ eV to get the same Curie temperature [18]. Surprisingly, although our calculations are done within a one-band model, this value agrees perfectly with the estimate $J = -1.1 \pm 0.2$ eV [19].

In Fig. 4 we analyze the dependence of T_C on the impurity concentration c , for different values of γ . For a given c , we observe that, T_C is non monotonic with respect to γ . However it is clear, that even at large concentration the low hole density is more favorable to get a high Curie temperature. More precisely T_C is larger when $\gamma \approx 0.1$. For instance, when $c = 0.15$ we get $T_C \approx 240$ K. Additionally, we see that when increasing c , for sufficiently large γ , T_C shows a maximum and decreases till it vanishes. It is expected that T_C will first vanish for larger itinerant carrier density. Indeed, the localization effect is stronger at higher carrier density.

To conclude, we have presented a theory to study ferromagnetism in DMS, which consists (i) in treating the itinerant carrier within the best single site approximation (CPA) and (ii) performing the susceptibility calculation with the disordered Green's functions to get the Curie temperature. We have shown that the role of disorder

is important and leads to significantly higher values of the Curie temperature with respect to a standard RKKY calculation. Additionally, it is shown that an antiferromagnetic coupling favors a higher T_C in the hole doped materials. We believe that this approach is more suitable to analyze ferromagnetism in diluted semiconductors.

One of us (J.K.) acknowledges the financial support provided by the Grant Agency of the Czech Republic (No. 202/00/0122) and the Grant Agency of the Academy of Sciences of the Czech Republic ((No. A1010203).

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 - [16] In the case of $S = 1/2$, this corresponds to a singlet state $E_S = -\frac{3}{4}J$ with degeneracy 1 and a triplet state $E_T = +\frac{1}{4}J$ with degeneracy 3.
 - [17] In GaAs the volume of the unit cell is $v = a_0^3/4$ where $a_0 = 0.565$ nm. For simplicity, our calculations are performed on a simple cubic lattice, thus the lattice spacing we take is $a_1 = (a_0^3/4)^{1/3}$.
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FIG. 1. Total DOS (left panels) and Mn projected DOS (right) as a function of $2E/W$ (bandwidth $W = 12t$), for majority spin (dashed curve) and minority spin (full curve) at $T = 0$ K. The value of J/t are $J/t = 3.45$ (a), $J/t = 2.33$ (b) and $J/t = 0.86$ (c). The concentration of impurity is $c = 5\%$.

FIG. 2. T_C/t as a function of $\gamma = n_h/c$ for different values of J/t . The concentration of impurity is fixed to $c = 5\%$.

FIG. 3. T_C/t as a function of J/t for different carrier density. The impurity concentration is $c = 5\%$. The RKKY calculation corresponds to the continuous, dashed and dotted curves and the full CPA treatment to symbols.

FIG. 4. T_C (in K) as a function of c for different values of γ (see fig.). The parameters are $t = 0.58$ eV and $J = 1.12$ eV. (see text)







